Rec'd PCT/PTO

DATE

AJZ:jvbp

IN THE UNITED STATES DESIGNATED/ELECTED OFFICE

420 Rec'd PCT/PTO 2 0 DEC 1999

INTERNATIONAL APPLICATION NO.

PCT/EP98/03250

INTERNATIONAL FILING DATE

30 May 1998

PRIORITY DATE CLAIMED

19 JUNE 1997

Applicant(s) (DO/EO/US)

Reiner DELP et al.

Title: LASER-MARKABLE PLASTICS

PRELIMINARY AMENDMENT

BOX PCT

Assistant Commissioner for Patents

Washington, D.C. 20231

SIR:

Please amend the above-identified International application as follows prior to calculation of the fees.

IN THE SPECIFICATION:

Page 2, line 19:

Delete "PE [sic]".

Page 5, line 10:

Delete "[sic]".

Page 6, line 13:

Delete "Vol. 15"

line 14:

Delete "[sic]".

Page 8, line 8:

Delete "[sic]".

Page 9, line 22:

Delete "biscuits [sic]" and insert -- platelets --.

Page 10, line 16:

Delete "biscuits [sic]" and insert -- platelets --.

Page 11, line 15:

Delete "methyl [sic]" and insert -- polymethyl--.

IN THE CLAIMS:

Please amend claims 3-6 and 8-11 as follows:

Claim 3, lines 1 and 2: Change "Claim 1 or 2" to -- Claim 1 --.

Claim 4, lines 1 and 2:

Change "one of Claims 1 to 3" to -- Claim 1 --.

Claim 5, lines 1 and 2:

Change "one of Claims 1 to 4" to -- Claim 1 --.

Claim 6, lines 1 and 2:

Change "one of Claims 1 to 5" to -- Claim 1 --.

Claim 8, lines 1 and 2: Change "one of Claims 1 to 7" to -- Claim 1 --.

Claim 9, lines 1 and 2: Change "one of Claims 1 to 8" to -- Claim 1 --.

line 5: Delete methyl [sic] and replace with polymethyl.

Claim 10, lines 1 and 2: Change "one of Claims 1 to 9" to -- Claim 1 --

REMARKS

The principal purpose of this Preliminary Amendment is to eliminate discrepancies in the text and to eliminate multiple dependencies in order to avoid extra fees.

Respectfully submitted,

Anthony J. Zelano (Reg. No. 27, 969)

Attorney for Applicants

MILLEN, WHITE, ZELANO & BRANIGAN, P.C.

Arlington Courthouse Plaza I

2200 Clarendon Boulevard, Suite 1400

Arlington, Virginia 22201 Direct Dial: 703-812-5311 Facsimile: 703-243-6410

Internet Address: zelano@mwzb.com

Filed: 20 December 1999

10

15

20

25

30

35

1 420 Rec'd PCT/PTO 2 0 DEC 1999

Laser-markable plastics

The present invention relates to laser-markable plastics of which a feature is that a plastic which is difficult to laser-treat comprises as absorber material one or more intrinsically laser-markable polymers in the form of micromilled particles having a particle size of 0.1 - 100 $\mu m.$

The labelling of production goods is becoming increasingly important in almost all sectors industry. For example, it is frequently necessary to apply production dates, expiry dates, barcodes, company logos, serial numbers, etc. At present, these marks are predominantly made using conventional techniques such printing, embossing, stamping and labelling. However, the importance of contactless, high-speed and flexible marking using lasers is increasing, especially in the case of plastics. This technique makes it possible to apply graphic inscriptions, for example barcodes, at high speed even on a non-planar surface. Since the inscription is within the plastics article itself, it is durable and resistant to abrasion.

Many plastics, for example polyethylene (PE), polypropylene (PP), polyamide (PA), polymethyl methacrylate (PMMA), polyoxymethylene (POM), polyurethane (PUR), polyesters have hitherto proved to be very difficult or even impossible to mark by means of the laser. A CO₂ laser which emits light in the infrared region at 10.6 µm produces only a faint, barely legible mark on polyolefins, even at very high output levels, since the absorption coefficient of the plastics to be processed is not high enough at these wavelengths to induce a colour change in the polymeric material. The plastic must not completely reflect or transmit the laser light, since if it did there would be interaction. However, there must also not be strong absorption, since in that case the plastic evaporates to leave only an engraving. The absorption of the laser beams and hence the interaction with the material

10

15

20

25

30

35

depends on the chemical structure of the plastic and on the laser wavelength employed. In many cases it is necessary to add appropriate additives, for example absorbers, in order to render plastics laserinscribable.

The article "Pearl Lustre Pigments - Characteristics and Functional Effects" in Speciality Chemicals, May 1982, Vol.2, No. 2 discloses the use of pearl lustre pigments for laser marking. Pearl lustre pigments, however, have the disadvantage that they severely alter the colour properties of the plastic, an effect which is often unwanted.

DE-A 29 36 926 discloses that the inscription of a polymeric material by means of laser light can be achieved by adding to the plastic a filler, such as carbon black or graphite, which discolours on exposure to energetic radiation.

EP 0 190 997 A prepares laser-inscribable moulding compounds, including PE, PE [sic] or PS, by adding at least one inorganic pigment to the high molecular mass organic material.

In EP 0 330 869, ${\rm TiO_2}$ and carbon black are added to PBT and PET. The inscription is dark on a light background. The use of carbon black and/or graphite as absorbers in connection with the laser marking of polyesters is known from EP 0 485 181.

However, the fillers known for laser marking have the disadvantage either that they permanently colour the plastic that is to be inscribed, and hence the laser inscription, which is usually a dark script on a lighter background, then lacks sufficient contrast, i.e. legibility, or that as in the case, for example, of kaolin the mark is very faint and becomes readily visible only when large amounts of the additive are employed.

DE 195 36 047 describes the use of polycarbonate, which itself is difficult to laser-mark owing to its amorphous structure, in a polymer matrix of a polyalkylene terephthalate. Through absorption of

20

30

35

laser energy it is possible for dark characters to be achieved on a light background in the polymer matrix of a polyalkylene terephthalate.

In addition to the abovementioned plastics, however, there are also polymers which can be marked by 5 means of a laser with darkness and great contrast without the addition of additives. Examples of such include polymers PET, butadiene-styrene polystyrene, polyphenyl ether (PPO), liquid crystal polymers (LCP), polyphenylene sulfide, polyarylates, 10 polyaryl sulfides, polyaryl sulfones, polyaryl ether ketones, and blends thereof.

The object of the present invention, therefore, was to find laser-markable plastics which permit high-contrast laser marking on exposure to laser light. In this context, the filler or successful absorber should have a very pale, neutral inherent colour and should possess the properties of the precoloured plastic that is to be marked or should have little or no effect on these properties.

It has surprisingly been found that a plastic which is difficult to laser-treat can be marked very effectively if, for example, one of the abovementioned intrinsically markable polymers is added in fine distribution to the plastic. The intrinsic markability of the polymer is thus transferred to the plastic which shows little if any such behaviour. Following laser bombardment, a plastic doped in this way shows high-contrast and crisply contoured markings even at low laser intensities.

The invention therefore provides laser-markable plastics, characterized in that plastics which are difficult to laser-treat comprise one or more intrinsically markable polymers having a particle size of from 0.1 to 100 $\mu m\,$

Through the addition of micromilled, intrinsically markable polymers as absorbers in concentrations of 0.1 to 10% by weight, preferably 0.1 to 5% by weight and, in particular, 0.1 to 2% by

10

15

20

25

30

35

weight, based on the plastics system, a high contrast is obtained in connection with laser marking. The concentration of the intrinsically markable polymers in the plastic, however, is dependent on the plastics system employed and on the laser used.

Suitable polymers or polymer mixtures are all known readily laser-treatable plastics, such as PET, ABS, polystyrene, PPO, polyphenylene sulfide, polyphenylene sulfone, polyimidosulfone and LCPs, for example.

Micromilled thermoplastics having a very high melting range of >300°C are particularly suitable. The crispness of the contours of the mark is determined in particular by the particle size of the micromilled polymers. The polymers preferably have particle sizes in the range from 0.1 to 50 μ m, in particular from 1 to 20 μ m.

The resulting mark is positively influenced if the intrinsically markable polymer comprises as further absorber, a light-sensitive pigment, such as a filler, for example, a conductive pigment and/or a special-effect pigment. The addition of a further absorber intensifies the contrast as a function of the plastics system used. The amount of light-sensitive pigment added should be between 0.1 and 90%

Particularly suitable light-sensitive pigments are fillers, such as TiO₂ and SiO₂, for example, and phyllosilicates. Suitable silicatic platelets here are, in particular, light-coloured or white micas. It is of course also possible to use other natural micas, such as phlogopite and biotite, synthetic mica, talc flakes and glass flakes. By special-effect pigments are meant all known lustre, metallic and pearl lustre pigments, as marketed, for example, by the companies Mearl, Eckart-Werken and Merck KGaA. Examples of suitable conductive pigments are the pigments marketed under the tradename Minatec® by Merck KGaA. These are platelet-shaped TiO₂/ mica pigments comprising an external tin/antimony oxide layer as conductive coat. Other

25

30

35

suitable light-sensitive pigments are the oxides, hydroxides, sulfides, sulfates and phosphates of metals such as copper, bismuth, tin, zinc, silver, antimony, manganese, iron, nickel and chromium, for example. Particular mention should be made in this case of the 5 use of antimony, bismuth oxychloride and copper(II) hydroxide phosphate. Particular preference in this context is given to a product as formed by heating blue Cu(II) orthophosphate (Cu₃(PO₄)₂·3 H₂O) heating [sic] to from 100 to 200°C, and which has the 10 'empirical chemical formula CuO.P.O.H.O Cu₃(PO₄)₂·Cu(OH)₂. Other suitable copper phosphates are 6 CuO.P.O.3 H_2O_1 , $Cu_3(PO_4)_2 \cdot 3$ $Cu(OH)_2$, 5 $CuO \cdot P_2O_5 \cdot 3$ $Cu_3(PO_4)_2 \cdot 2$ $Cu(OH)_2 \cdot H_2O_1$ 4 $CuO \cdot P_2O_5$ 4 $CuO \cdot P_2O_5 \cdot 3H_2O_7$ $CuO \cdot P_2O_5 \cdot 1.5 H_2O$, 4 $CuO \cdot P_2O_5 \cdot 1.2 H_2O$. 15

An improvement in laser markability is also achieved if, in addition to the intrinsically marking polymer, one or more abovementioned light-sensitive pigments are added as a further component to the plastic. In this case the proportion by weight of all absorbers in the plastic, in combination with the micromilled polymers, should not exceed a total of 10% by weight, based on the plastics system. The plastic preferably contains 0-5% by weight of light-sensitive pigments, especially 0-1% by weight. In this context, there is no restriction on the proportion in which the light-sensitive pigments are mixed with the micromilled polymers.

It is also possible to add, to the plastic that is difficult to laser-treat, colour pigments, which permit colour variations of any type and at the same time ensure that the laser marking is retained.

The light-sensitive pigments and/or colour pigments are added preferably together with the polymers, although separate addition is also possible in principle. A mixture of different light-sensitive pigments can also be added to the plastic.

Marking is preferably carried out using highenergy radiation, generally in the wavelength range

15

20

25

from 150 nm to 10,600 nm, in particular in the range from 150 nm to 1100 nm. Mention may be made here, for example, of CO_2 lasers (10,600 nm), Nd:YAG lasers (1064 nm or 532 nm) or pulsed UV lasers (excimer lasers). Particular preference is given to the use of Nd:YAG lasers (1064 nm and 532 nm) and CO_2 lasers (10,600 nm). The energy densities of the lasers employed are generally in the range from 0.3 mJ/cm² to 10 J/cm^2 , preferably in the range from 0.3 mJ/cm² to 10 J/cm^2 .

All known plastics which can only be lasermarked with great difficulty, as are described, for example, in Ullmann, Vol. 15, p. 457 ff., Vol. 15 [sic], Verlag VCH or Saechtling Kunststoff Taschenbuch, can be employed for laser marking through addition of polymers of the invention. Examples of plastics are thermosets, polyethylene (PE-HD, PE-LD, PE-LLD), polypropylene (PP), polyesters, polyacetal, (PA), polyurethanes polyamides (PUR), polybutylene polymethyl terephthalate, methacrylate polyvinyl acetal, polystyrene, butadiene-styrene (ABS), acrylonitrile-styrene-acrylate (ASA), and copolymers and/or mixtures thereof. In particular, polyolefins, polyurethanes, polyoxymethylenes polyamides, owing to their mechanical properties, the cost-effective processing methods and their poor lasermarkability, are suitable for doping with the polymers of the invention.

The incorporation of the micromilled polymer into the plastic takes place by the techniques known for pigments and fillers. Subsequently, the pigmented plastic is then deformed under the action of heat. When choosing the intrinsically markable polymer to be milled it should be borne in mind that the particle structure is retained following the incorporation; in other words, the particles should not be soluble in the melt, and should not melt as well. This is achieved by appropriate tailoring of the melting ranges of the plastics system to that of the micromilled polymer.

15

20

25

30

35

When the micromilled polymer is incorporated into the plastics granules it is possible, if desired, to add coupling agents, organic, polymer-compatible solvents, stabilizers, optical brighteners, pigments, fillers, reinforcing dyes, agents, flameproofing additives, antistatics and/or surfactants which are temperature-stable under the operating conditions. In addition to the auxiliaries customarily employed it is possible to add further additives, not mentioned here, to the plastic. The presence of further additives in the existing plastics systems, however, may exert an effect on the marking result.

The plastics granule/polymer mixture is generally prepared by charging an appropriate mixer with the plastics granules, wetting them with any additives, and then adding the micromilled polymers and mixing them in. The mixture obtained in this way can then be processed directly in an extruder or an injection moulding machine. The mouldings formed in the course of processing usually exhibit a very homogeneous distribution of the polymers or of the polymer mixture. Finally, laser marking takes place, preferably with a Nd:YAG laser.

Inscription with the laser is carried out by introducing the sample into the beam path of a pulsed laser, preferably a Nd:YAG laser. Inscription with a CO₂ laser or an excimer laser is also possible. However, the desired results can also be achieved with other types of laser featuring a wavelength in a range of high absorption by the intrinsically marking polymer. The resulting shade and depth of colour are determined by the laser parameters, such as the irradiation time and irradiation output. The output of the lasers used depends on the particular application and can be determined regularly in each individual case by the skilled worker.

The plastic doped in accordance with the invention can be used in all sectors where customary printing processes have hitherto been employed for the

inscription of plastics. For example, mouldings of the plastic of the invention can be employed in electrical, electronic and motor vehicle industry. The labelling and inscription of, for example, housings, lines, keycaps, transcripts or functional parts in the heating, ventilation and cooling sectors, or switches, plugs, levers and handles which consist of the plastic of the invention, can be marked [sic] with the aid of laser light, even at difficult-to-reach points. Owing to its low heavy-metal content, the plastics system of 10 the invention can also be employed in packaging in the foodstuffs sector or in the toy sector. The markings on packaging are notable for their resistance to wiping and scratching, for their stability during subsequent sterilization processes, and for the fact that they can 15 be applied in a hygienically pure manner in the marking process. Complete label motifs can be applied durably the packaging for a reusable system. important area of application for laser inscription is 20 that of identity cards and plastic tags for the individual identification of animals: so-called cattle The laser marking of plastics or earmarks. articles or mouldings which consist of the plastic of the invention is hence possible.

The examples which follow are intended to illustrate the invention without, however, restricting it.

Examples

Example 1

5 99 parts of polypropylene (Stamylan PPH 10 from DSM)
1 part of polyphenylene sulfide milled to a particle
size < 25 μm

The components are physically mixed and by means of an injection moulding machine are homogenized and shaped to form platelets. The inscription with a 'Nd:YAG laser at 532 and 1064 nm wavelengths shows a high-contrast black marking with a smooth surface over a wide range of settings.

15 <u>Example 2</u>

99.5 parts of polypropylene (Stamylan PPH 10) 0.5 part of polyphenylene sulfone milled to a particle size < 10 μm

The components are mixed and by means of an injection moulding machine are homogenized and shaped to form biscuits [sic]. The inscription with a Nd:YAG laser shows a high-contrast black marking with a smooth surface over a wide range of settings.

Erramala :

25

35

Example 3

99 parts of polyamide 6 (Ultramid B3K from BASF)

1 part of polyimidosulfone milled to a particle size

 $< 15 \mu m$

The components are mixed and by means of an injection moulding machine are homogenized and shaped to form platelets. The inscription with a Nd:YAG laser shows a high-contrast deep-black marking with a smooth surface over a wide range of settings.

Example 4

99.6 parts of polyamide 6 (Ultramid B3K)

0.4 part of polyphenylene sulfide milled to a particle size < 10 μm

The components are mixed and by means of an injection moulding machine are homogenized and shaped to form platelets. The inscription with a Nd:YAG laser shows a high-contrast black marking with a smooth surface over a wide range of settings.

Example 5

10

25

30

199 parts of polyoxymethylene (Delrin from Du Pont)

1 part of polyphenylene sulfide milled to a particle size < 5 µm

The components are mixed and by means of an injection moulding machine are homogenized and shaped to form biscuits [sic]. The inscription with a Nd:YAG laser shows a high-contrast black marking with a smooth surface over a wide range of settings.

20 Example 6

99 parts of unsaturated polyester resin (Palatal from BASF)

1 part of polyphenylene sulfide milled to a particle size < 10 μm

The polyphenylene sulfide is incorporated homogeneously into the liquid polyester casting resin by stirring. Following the addition of accelerator (Co octoate) and hardener (cyclohexanone peroxide), the mixture is poured into a mould. After curing has taken place, a moulding is obtained which can be given a high-contrast black marking by means of Nd:YAG lasers.

Example 7

99 parts of polysulfone (Ultrason from BASF) are compounded on an extruder together with 1 part of mica. The compound is micromilled to a particle size of $<10~\mu m.$ 0.5% of the powder thus obtained are added to a PMMA. This mixture is processed on an extruder to

give sheets which can be given a black and high-contrast marking with a Nd:YAG laser at a wavelength of 532 and 1,064 nm.

5 Example 8

10

96 parts of polyphenylene sulfide are compounded by the method of Example 7 with 4 parts of basic copper phosphate. An addition of just 0.4% of the micromilled powder of this mixture to commonly non-laser-markable plastics, such as.

- (a) polyethylene (PE)
- (b) polypropylene (PP)
- (c) polyamide (PA)
- 15 (d) methyl [sic] methacrylate (PMMA)
 - (e) polyurethane (PU)
 - (f) polyoxymethylene (POM)

gives deep-black, high-contrast markings with crisp contours using a Nd:YAG laser.

Patent Claims

- 1. Laser-markable plastics, characterized in that a plastic which is difficult to laser-mark comprises as absorber material an intrinsically laser-markable polymer in the form of micromilled particles having a particle size of 0.1 100 μm .
- Laser-markable plastics according to Claim 1,
 characterized in that the absorber material is a high-temperature-resistant plastic.
 - 3. Laser-markable plastics according to Claim 1 or 2, characterized in that the absorber material is polyphenylene sulfide, polysulfone, polyarylate,
- 15 polyimide, liquid-crystalline polymers or a mixture thereof.
 - 4. Laser-markable plastics according to one of Claims 1 to 3, characterized in that the proportion of the absorber material based on the plastics system is 0.1 10% by weight.
 - 5. Laser-markable plastics according to one of Claims 1 to 4, characterized in that the particle structure of the intrinsically markable polymer is retained in the plastic.
- 25 6. Laser-markable plastics according to one of Claims 1 to 5, characterized in that the absorber material additionally comprises, as further absorber, one or more light-sensitive pigments.
- 7. Laser-markable plastics according to Claim 6,
 30 characterized in that the light-sensitive pigment is
 natural or synthetic mica, copper phosphate, a specialeffect pigment, a conductive pigment, a metal nitrate,
 metal sulfate, metal sulfide or metal oxide.
- 8. Laser-markable plastics according to one of Claims 1 to 7, characterized in that the proportion of the light-sensitive pigments in the plastic is from 0 to 5% by weight, based on the plastics system.
 - 9. Laser-markable plastics according to one of Claims 1 to 8, characterized in that the plastic which

- is difficult to laser-treat is polyethylene, polypropylene, polyamide, polyoxymethylene, polyester, methyl [sic] methacrylate, polyurethane or a copolymer thereof.
- 5 10. Laser-markable plastics according to one of Claims 1 to 9, characterized in that they additionally comprise colour pigments.
 - 11. Use of the laser-markable plastics according to Claim 1 as material for producing mouldings which are marked with the aid of lasers.
 - 12. Mouldings consisting of the laser-markable plastic according to Claim 1.

Abstract

The present invention relates to laser-markable plastics of which a feature is that a plastic which is difficult to laser-treat comprises as absorber material an intrinsically laser-markable polymer in the form of micromilled particles having a particle size of 0.1 - $100~\mu m$.

Docket No. **MERCK**

Declaration and Power of Attorney For Patent Application

English Language Declaration

As a below named inventor, I hereby declare that:

My residence, nost office address and citizenship are as stated below next to my name

which a patent is sou			~
Laser-markable pla	astics		
the specification of wh	hich		
(check one)			
☐ is attached hereto	Э.		
🗵 was filed on May	⁷ 30, 1998 a	as United States Application No.	or PCT International
Application Numb	per PCT/EP 98/03	250	
and was amended	d on		
		(if applicable)	
l acknowledge the du	uty to disclose to the Unite	Iment referred to above. d States Patent and Trademark	
I acknowledge the duknown to me to be Section 1.56. I hereby claim foreig Section 365(b) of an any PCT International listed below and have inventor's certificate of	uty to disclose to the Unite material to patentability a gn priority benefits under by foreign application(s) for all application which designate also identified below, by or PCT International applic		Section 119(a)-(d) of the control of
I acknowledge the duknown to me to be Section 1.56. I hereby claim foreig Section 365(b) of an any PCT International listed below and have	uty to disclose to the Unite material to patentability a gn priority benefits under by foreign application(s) for all application which designate also identified below, by or PCT International application.	d States Patent and Trademarks defined in Title 37, Code of Title 35, United States Code, patent or inventor's certificate ated at least one country other to checking the box, any foreign a	Section 119(a)-(d) of the control of
I acknowledge the duknown to me to be Section 1.56. I hereby claim foreig Section 365(b) of an any PCT International listed below and have inventor's certificate on which priority is class.	uty to disclose to the Unite material to patentability a gn priority benefits under by foreign application(s) for all application which designate also identified below, by or PCT International application.	d States Patent and Trademarks defined in Title 37, Code of Title 35, United States Code, patent or inventor's certificate ated at least one country other to checking the box, any foreign a	Section 119(a)-(d) of section 119(a)-(d) of section 365(a) of han the United States pplication for patent of that of the application
I acknowledge the duknown to me to be Section 1.56. I hereby claim foreig Section 365(b) of an any PCT International listed below and have inventor's certificate con which priority is claim.	uty to disclose to the Unite material to patentability a gn priority benefits under my foreign application(s) for all application which designate also identified below, by or PCT International application(s)	d States Patent and Trademarks defined in Title 37, Code of Title 35, United States Code, patent or inventor's certificate ated at least one country other to checking the box, any foreign a ation having a filing date before	Section 119(a)-(d) of section 365(a) of han the United States pplication for patent of that of the application Priority Not Claimed
I acknowledge the duknown to me to be Section 1.56. I hereby claim foreign Section 365(b) of an any PCT International listed below and have inventor's certificate of on which priority is claim. Prior Foreign Application DE 197 26 136.1 (Number)	uty to disclose to the Unite material to patentability a gn priority benefits under my foreign application(s) for all application which designate also identified below, by or PCT International application(s) Cermany (Country)	d States Patent and Trademarks defined in Title 37, Code of Title 35, United States Code, repatent or inventor's certificate ated at least one country other to checking the box, any foreign a ation having a filing date before 19 June 1997 (Day/Month/Year Filed)	Section 119(a)-(d) of section 365(a) of han the United States pplication for patent of that of the application
I acknowledge the duknown to me to be Section 1.56. I hereby claim foreig Section 365(b) of an any PCT International listed below and have inventor's certificate on which priority is claim Prior Foreign Applicated DE 197 26 136.1	uty to disclose to the Unite material to patentability a gn priority benefits under by foreign application(s) for all application which designate also identified below, by or PCT International application(s)	d States Patent and Trademarks defined in Title 37, Code of Title 35, United States Code, repatent or inventor's certificate ated at least one country other to checking the box, any foreign a ation having a filing date before	Section 119(a)-(d) of section 365(a) of han the United States pplication for patent of that of the application Priority Not Claimed

(Application Serial No.)	(Filing Date)	
(Application Serial No.)	(Filing Date)	
(Application Serial No.)	(Filing Date)	
nsofar as the subject matter of ea United States or PCT International U.S.C. Section 112. I acknowledge	ach of the claims of this app application in the manner p to the duty to disclose to the U	lication is not disclosed in the prior rovided by the first paragraph of 35 Jnited States Patent and Trademark
nsofar as the subject matter of ea United States or PCT International U.S.C. Section 112. I acknowledge Office all information known to me	ach of the claims of this app application in the manner po to the duty to disclose to the U to be material to patentabi alle between the filing date of the	lication is not disclosed in the prior rovided by the first paragraph of 35 Jnited States Patent and Trademark lity as defined in Title 37, C. F. R.,
nsofar as the subject matter of ear United States or PCT International U.S.C. Section 112. I acknowledge Office all information known to me Section 1.56 which became availab	ach of the claims of this app application in the manner po to the duty to disclose to the U to be material to patentabi alle between the filing date of the	lication is not disclosed in the prior rovided by the first paragraph of 35 Jnited States Patent and Trademark lity as defined in Title 37, C. F. R.,
Insofar as the subject matter of ear United States or PCT International U.S.C. Section 112. I acknowledge Office all information known to me Section 1.56 which became availab or PCT International filing date of th	ach of the claims of this app application in the manner parties the duty to disclose to the less to be material to patentable between the filing date of this application:	

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

	application and transaction and registration numes (544) (6) (7,969) (20,565) (3) (004) (542)	I hereby appoint the following that all business in the Patent anber) Diana Hamlet-King (Reg. No. 33,103) John A. Sopp (Reg. No. 33,103) Richard E. Kurtz (Reg. No. 33,9 Richard M. Lebovitz (Reg. No. 33,46) John H. Thomas (Reg. No. 33,46)	and Trademark Office 3,302) 36) 37,067)
Send Correspondence to: Direct Telephone Calls to:	MILLEN, WHITE, ZELA Arlington Courthouse Pla 2200 Clarendon Blvd., Su Arlington, VA 22201 (name and telephone no	za I	
Full name of sole or first inventor			
Reiner Delp Sole of first inventors		Septeml	Date Oer 6, 1999
Residence	ach 64193 Darmst	16	
64407 Fränkisch-Crumb	ach 67173 Jaimst	<u> </u>	
Citizenship German			
Post Office Address			
c/o Merck KGaA, 64271	Darmstadt/Germany	DEX	
Full name of second inventor, if	any		
Jürgen Solms	,		
Second inventor's signature		Comtomb	Date
J. James		Septem	per 6, 1999
Hesidence 64319 Pfungstadt			
Citizenship			
German			

Post Office Address

c/o Merck KGaA, 64271 Darmstadt/Germany

Third hventoris signature		Date
Third inventoris signature	September 6, 1999	
Residence 55468_Trebur		
Citizenship		
German Post Office Address		
Post Office Address C/o Merck KGaA, 64271 Darmstadt/Germany	ラ人	
Full name of fourth inventor, if any		
Fourth inventor's signature		Date
Residence		
Cıtizenship		
Post Office Address		
Full name of fifth inventor, if any		
Full name of fifth inventor, if any Fifth inventor's signature		Date
		Date
Fifth inventor's signature		Date
Fifth inventor's signature Residence		Date
Fifth inventor's signature Residence Citizenship		Date
Fifth inventor's signature Residence Citizenship		Date
Fifth inventor's signature Residence Citizenship		Date
Fifth inventor's signature Residence Citizenship		Date
Fifth inventor's signature Residence Citizenship Post Office Address		Date
Fifth inventor's signature Residence Citizenship Post Office Address Full name of sixth inventor, if any		
Fifth inventor's signature Residence Citizenship Post Office Address Full name of sixth inventor, if any Sixth inventor's signature		
Fifth inventor's signature Residence Citizenship Post Office Address Full name of sixth inventor, if any Sixth inventor's signature Residence		